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Use of Lasers to Study the Impact of Fractionation and Condensation on the Toxicity of Nuclear Weapon Fallout

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Abstract:

An experimental concept has been developed to collect data to aid in the refinement of simulation programs designed to predict the fallout effects arising from surface and shallowly buried nuclear weapon detonations. These experiments, called the Condensation Debris Experiments (CDE), are intended to study the condensation/fractionation of material that is liberated following an initial deposition of laser energy onto a small, characterized target. The CDE effort also encompasses target development and material studies as well as supporting computational efforts studying radiation hydrodynamics, computational fluid dynamics, and relevant neutron activation processes (not discussed here).

Introduction:

During the atmospheric testing era, a large effort was made to measure the gamma activity as a function of position and time, both near (m) and far (km) from ground zero [1]. Although efforts to determine the activity contours were made for all tests, the manner in which the majority of the data was collected was not precise. In order to quantify the uncertainty that exists in the data that was collected during the atmospheric testing era, data from Shot Owens of operation Plumbbob was analyzed [2]. Owens was a 9.7 kt device detonated at a height of 500 ft [1]. The gamma activity pattern generated from the shot is primarily due to neutron-induced activity. Readings were taken by hand along 8 radial roads converging at ground zero. Data from two of those roads (94 and 204 degrees) are unclassified and available. The surveys occurred at H+1 hour, H+6 hours, D+1, D+2, D+3, and D+5 days. The readings that were recorded varied from 0.06 to 17.5 rem/hr. The uncertainty associated with the measurement technique was quantified by determining the standard deviation of the measurements, which is 16%. While this level of uncertainty was sufficient to determine when a region near where a nuclear burst occurred was safe for military personnel to return in the 1960s, the data are simply not accurate enough to assist in the validation of contemporary codes that are tasked with predicting more subtle effects, like total radiation dose on a population. In order to model this type of effect, the accuracy of the experimental data needs to be better than 5%.

In addition to the gamma activity surveys, other analyses discovered an important phenomenon, fractionation. Fractionation is the process by which the concentrations of radioactive elements created and/or dispersed by a nuclear weapon varies from the initial

concentrations. Fractionation is generally caused by the intrinsic physical properties of the materials present in the radioactive cloud during the condensation process. Materials with the highest boiling points, the refractory elements, will condense first, leaving a higher relative concentration of the most volatile elements in the radioactive cloud. The degree of fractionation is generally a function of the height of burst. This is due to the amount of atmospheric and ground material present in the fireball at the condensation time. (Atmospheric and ground material in the fireball constitutes a significant heat sink, which allows the material in the fireball to cool at a faster rate, enhancing fractionation and the condensation rate.)

As mentioned, the work that was done in the atmospheric testing era to measure and characterize the fallout that was generated by nuclear detonations was successful in determining when it was relatively safe for humans to reenter the area where the detonation occurred. In addition enough data were collected and analyzed in order to begin generating models that predicted the fallout from specific nuclear bursts to within a factor of 2 or 3 of the measured rate [3]. However, the collection methods, analysis methods, and models were not flexible or precise enough to measure and predict fallout phenomena with the accuracy required for today's applications. The Condensation Debris Experiment has been developed to fill the gaps in the understanding of nuclear fallout and help provide the flexibility that is necessary to develop and test high precision fallout models.

Experimental Setup:

An experimental platform that allows the study of the physical processes and various empirical laws relating to fallout will lead to an improved understanding of the entire fallout generation process. The approach of the CDE effort is to create a plasma by depositing laser energy into a small high-Z laser target (a 'halfraum'), converting the laser energy into x-rays and debris. The choice of a laser for the energy source has substantial benefits when compared to other energy sources, most notably high explosives (HE). Lasers can generate plasmas with energy densities that are relevant when compared to those created in nuclear explosions. This is due to the large amount of energy (10s of kJ) that contemporary laser facilities can generate in short pulse lengths (ns) combined with flexibility in the target design, minimizing both physical size and mass. Tailoring of the target and laser pulse allows experimenters to modify the x-ray and debris output, providing a versatile experimental platform for nuclear weapons effects studies. In addition, selecting a laser to supply the energy eliminates non-condensable gases that accompany HE charges.

In order to utilize the laser, two holes were drilled into a "soil surrogate." (The "soil surrogate has an elemental composition similar to soil or rock.) The exit shaft was drilled from the surface of the soil surrogate to the target area, representing an idealized path of a penetrating weapon. The laser entrance cone was drilled perpendicular to the exit shaft. The cone's area was at least an order of magnitude smaller than the exit shaft area allowing the debris to expand preferentially into the exit shaft. In addition to maintaining a large ratio of the two areas, the x-rays emitted from the halfraum laser entrance hole

(LEH) provides an additional protection against debris passing up the laser beam line by ablating thin layers of the soil surrogate, which then expand and temporarily close off the beam line.

Parameter	CDE	Nuclear Weapon
Plasma temp.	10^6 K	10^8 K
Plasma volume	1 mm^3	$10\text{-}100 \text{ m}^3$
Entrained mass	1 mg	$10^{12}\text{-}10^{15}$ mg
Energy released	$10^3\text{-}10^6$ J	$10^{12}\text{-}10^{15}$ J
# of neutrons	0	$10^{23}\text{-}10^{24}$
Initial debris mass	1-10 mg	1-100 kg
X-ray fraction	0-50 %	70-80%
Peak shock pressure	10 kbar	1 Gbar
Condensation time	<1s	20-25s

Table 1: A comparison of orders of magnitude for different experimental parameters for the CDE and nuclear weapons.

When the laser energy is deposited, it interacts with the back wall of the halfraum, generating x-rays. In addition, some x-rays escape through the LEH of the halfraum and impinge on the inner surface of the soil surrogate, ablating some material. After a short time, on the order of ns, the halfraum explodes, allowing the remaining x-rays and debris to expand and interact with the inner walls of the soil surrogate. The interaction of the x-rays and debris with the walls of the soil surrogate ablates additional material from the walls and launches large shocks (10's of kbars) that cause substantial local fracturing and pulverization of the soil material. This material expands through the exit shaft, escaping into a region where condensation/fractionation takes place.

The debris emanating from the exit shaft was contained in a vessel called the condensation chamber. The four main purposes the condensation chamber served were, to prevent the debris from entering the laser's target chamber, allow the debris to expand and condense, allow *in situ* diagnostics to characterize the debris cloud, and collect the debris so it could be studied after the experiment. The condensation chamber is cylindrical in shape. The determination of the height and diameter of the chamber was made with the assistance of the hydrodynamics code TSUNAMI, developed at the University of California [4]. The diameter and height of the condensation chamber were maximized in order to ensure the highest probability of in-flight condensation. The limitation for the size of the condensation chamber was the size of the vacuum chamber at the Z-Beamlet Laser facility. The final internal dimensions of the condensation chamber were 5¼ in. in diameter and 4.5-8 in. long. The condensation chamber was also designed to maintain chamber pressures larger than that of the surrounding vacuum chamber. O-rings and a thin MYLAR (~10 µm) membrane were utilized to seal the chamber. Finally, a clear cylindrical piece of acrylic was chosen to be the principle material for the cylindrical walls of the condensation chamber. The choice of clear acrylic allowed CCD images to be taken during the experiment.

Located below the condensation chamber was the soil containment vessel, designed to assure that if the soil surrogate disassembled, no shrapnel or substantial debris would escape. The containment vessel was cylindrical, 6 inches in diameter with 1 inch thick walls and was machined from a single piece of 6061 aluminum. The inner length of the soil containment vessel was 4 inches with a 1-inch thick base. Holes were machined in the containment vessel to allow the laser to pass into the soil surrogate as well as for placement of diagnostics. The soil surrogate was inserted into a 4 inch outside diameter, thin walled stainless steel container, within the soil containment vessel. This additional container assured that the soil surrogate could be potted with epoxy if needed, without rendering the aluminum containment vessel useless. In addition, the soil surrogate could be shock isolated within the stainless steel container. The soil containment vessel and the condensation chamber were bolted together to further assure structural integrity.

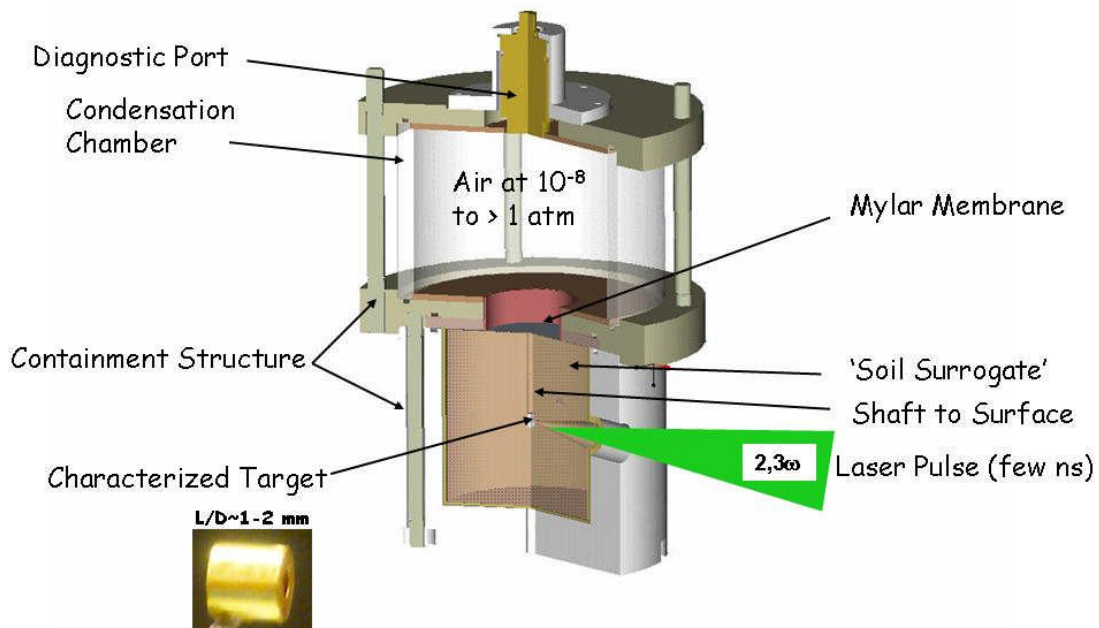


Figure 1: A cut away drawing of the experimental apparatus used in the Condensation Debris Experiment. The lower half of the apparatus consists of the soil surrogate, with the target imbedded inside, and its containment vessel. The upper half of the apparatus consists of the condensation chamber with an optional dynamic pressure sensor diagnostic attached to the very top of the chamber.

Several halfraum designs have been used throughout the course of the various CDE experimental campaigns. Variations in the halfraum design have included the size of the halfraum, the wall thickness, the internal material, and the wall construction. Ongoing collaborations with Atomic Weapons Establishment (Aldermaston, UK) and The Schafer Corporation (Livermore, CA) allow development and manufacturing of halfraum targets tailored to specific needs. Varying the size of the halfraum changes the energy density and the internal halfraum temperature. In addition to the internal temperature and energy

density, the amount of x-rays escaping the halfraum can be adjusted by varying the LEH diameter, the wall thickness, and/or the halfraum construction technique.

Halfraum Type	Length (mm)	Diameter (mm)	Wall Thickness (μm)	LEH Diameter (mm)	Internal Material?	Vendor
1	2.5	2.5	25	500	no	AWE
2	2.2	2.2	25	500	no	AWE
3	1	1	25	300	no	AWE
4	1	1	25	400	No	AWE
5	2	2	3	400	Doped foam	Schafer
6	2	2	3	400	Doped foam	Schafer

Table 2: Listing of the halfraums designed and tested in the CDE.

Modeling the Experimental Apparatus:

The various time and length scales of the experiment allow for the use of different models to simulate the different phases of CDE. The first set of models relied on are radiation-hydrodynamics codes that study the timeframe from the initial deposition of the laser energy through the disassembly of the halfraum. The radiation-hydrodynamics models that are being used in this stage include LASNEX [5] and HYDRA [6], both developed at Livermore (LLNL). At this time, the CDE source models are taking advantage of the cylindrical symmetry of the halfraum target and are only modeling the source in 2 dimensions. The outputs of these models are compared with experimental results including the internal temperature of the halfraum, the momentum distribution of the halfraum debris as a function of position, and the x-ray spectrum as a function of position and time. These predictions are then used as input into the second stage of CDE modeling.

The second stage is simulated through the use of thermal-mechanical models to study the interactions of x-rays and debris with the walls of the soil surrogate. To understand the x-ray interaction with the walls, a model is needed that predicts the amount of material ablated from the soil surrogate walls and the amount of material that is crushed due to the impact of the x-rays. These models also have the capability to predict the magnitude and the time dependence of the shock that is induced into the soil surrogate. LASNEX and HYDRA can be employed but further work is required to properly model the complex phase change of the soil surrogate. There are a number of potential models for simulating this phase change, including ALE3D [7].

The third modeling stage for the CDE involves gas dynamics and aerosol formation and transport models. These models simulate the flow of the debris cloud as it expands through the drift and into the condensation chamber. The outputs from these models include density, pressure, and temperature as a function of position and time. TSUNAMI is being utilized for this study. As a first cut to the third stage, hydrodynamics runs have been performed, but more comprehensive simulations are planned. A condensation package is under development for TSUNAMI that will help predict the particle size distribution.

As explained earlier, the results of the CDE are to be used to assist in the validation of codes that predict the fallout hazard from nuclear burst. In order to generate predictions for full-scale nuclear fallout hazard, a number of different numerical models are utilized. The first model type, as is used for modeling the first and second stage of CDE, is a radiation hydrodynamics model that generates the x-ray source term for the explosion and simulates the growth of the fireball until hydrodynamic separation. This model generally simulates the relevant processes that are taking place up to ~ 1 second after detonation. During the so-called blastwave phase, up to ~ 3 s, the growth of the fireball is governed by hydrodynamics and is simulated at LLNL with a compressible code AMR [8]. The third stage in the fallout modeling effort is called the cloud rise phase. This phase lasts from ~ 3 to 120s after detonation and is the focus of a large portion of the effort for improving the fallout predictive capability. The fourth phase, the cloud dispersion phase, utilizes atmospheric transport models to disperse the fallout particles across real world terrain for real world weather conditions. After the cloud dispersion phase is completed, predictions of the biological hazard as a function of position and time are determined.

With the resumption of nuclear testing unlikely, the best possible source for new data is through other experimental avenues. The CDE is one of these avenues. Information garnered from the CDE effort will allow an additional database for these new codes to compare their predictions against. The data that is most interesting to the nuclear fallout modeling community includes the particle size distribution and the spatial and time dependence of the debris cloud's pressure, temperature, density, and composition. In addition, the nuclear fallout modeling community will benefit through the refinements made on the radiation-hydrodynamics models through the study of the CDE source term.

Data Analysis:

During the November 2003, 11 CDE experiments were performed at the ZBL facility at Sandia National Laboratory in Albuquerque, NM, utilizing a brass soil surrogate and a bisected, non-pressurized condensation chamber. Three of these experiments were performed in order to determine the dynamic pressure history a set distance (10 cm) from the center of the halfraum. The measurements were performed with two types of pressure sensors manufactured by KTECH Corporation (Albuquerque, NM). The first pressure sensor used was a quartz crystal device, while the other two sensors used were plastic devices called PVDFs.

As can be seen in the long-exposure CCD image in figure 2, hot material exiting the soil surrogate cools as it expands, reaching hydrodynamic conditions where the debris' radiation no longer is sufficient to be detected. The radiation becomes visible again when the debris strikes the pressure sensor, where it stagnates and heats. This effect is reproduced nicely by the TSUNAMI code (showing the same effect for the vapor pressure), also seen in figure 2.

There were several technologies used to diagnose the November 2003 experiments. The NIKON NEXIV microscope was utilized for pre and post-shot metrology of the soil

surrogate. This determined the amount of material removed from the lower hemisphere of the target cavity. The average amount of material removed from the lower hemisphere was 109 ± 27 mg, for an average 2ω laser energy of 1.381 ± 0.146 kJ. Figure 3 shows an example of one of the NEXIV scans utilized in the study.

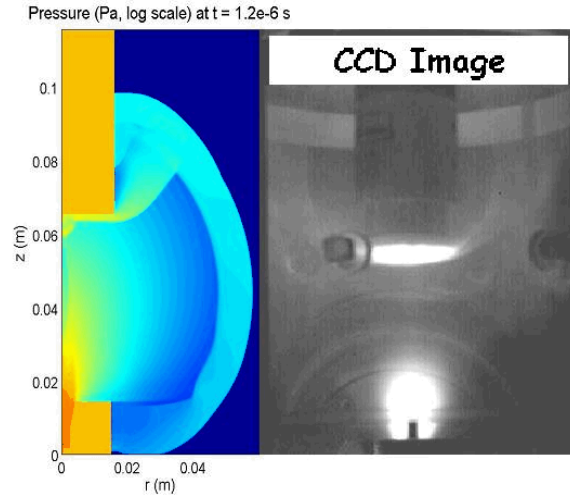


Figure 2: A snapshot of a TSUNAMI calculation (left) with a CCD image taken for one of the PVDF shots (right).

X-ray fluorescence (XRF) was used to determine the composition of the material deposited on the principal collection surfaces as a function of position. The upper and lower principal collection surfaces were analyzed at 15 different positions on each plate. At each scan point, a 40 mA x-ray beam (2 mm in diameter) impinged onto the collection surface, causing the deposited material on the surface to fluoresce. The photons that were released by the elements at characteristic energies were counted. The partial results of this study can be seen in figure 4.



Figure 3: Photos of a CDE surface experiment before (left) and after (center) laser deposition and a NEXIV microscope scan of the post-shot cavity (right).

The most important effect seen in the XRF study is that the elemental composition of the deposited material varies greatly across the sample. The composition of the brass that was used in these CDE experiments consisted of nearly 65% copper to 35% zinc, a zinc to copper ratio of 0.53. This homogenous mixture of the two elements was not reproduced on the collection surfaces. The relative amount of zinc to copper varied from 0.35 to 1.44. This is direct evidence of fractionation in the CDE experiments.

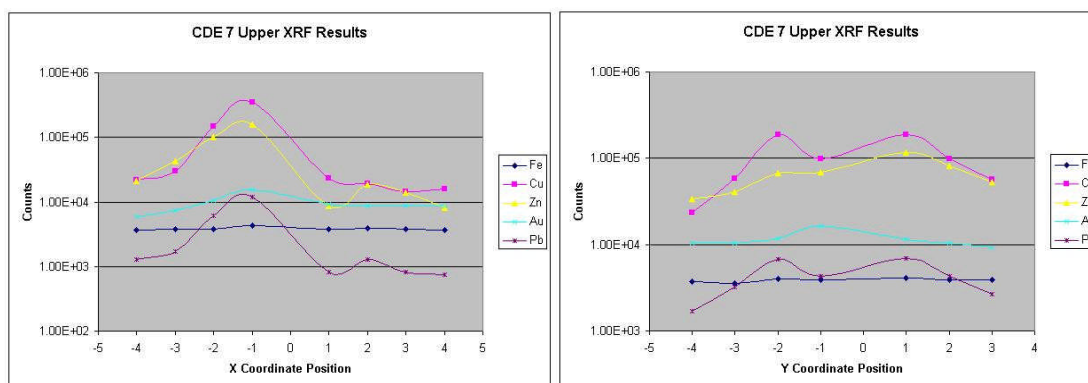


Figure 4: XRF elemental counts as a function of position taken for Nov. 2003 CDE shot #7 (the uncertainty in the results are within the data points). Results are for two orthogonal, bisecting lines defined as the x (left) and y (right) axes of the upper collection surface. Note: the gold composition remains fairly constant while the copper and zinc compositions vary both generally and relatively.

In addition to the NEXIV microscope and XRF, other technologies were utilized to collect data and study the CDE. Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) was utilized to measure the amount of liberated material on each collection surface and showed that up to 40% of the target mass could escape through the voids in the bisected condensation chamber. This result facilitated a redesign of the condensation chamber to that shown in figure 1. Finally, to study the composition of individual (condensed) particles, Scanning Electron Microscopy (SEM) was utilized. The results of the spectroscopic SEM study supports the XRF fractionation data by showing variations in the zinc to copper ratio of up to a factor of two on a particle-by-particle basis.

Conclusion:

The Condensation Debris Experiment is a laser based experimental platform that has been developed, utilizing cutting edge technology, in order to study the effects of fractionation and condensation on nuclear weapon fallout. To that end, a series of experiments have been performed on the Z-Beamlet Laser. These experiments have shown that a laser can be utilized as an energy source for creating a small and controlled "fallout" experiments. These preliminary experiments were also able to detect fractionation, an important component in understanding fallout formation. Work continues to improve these experiments with the goal of developing an empirical database that will be used to assist in validating first principle nuclear fallout models, such as the low-Mach number AMR code that is being used to study radioactive cloud rise. This work was performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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